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This project started on April 1, 2000 and was completed on April 30, 2003. The overall goal of the project is to develop novel processes, such as the cation exchange process, for epitaxy of 1-Ig-based high temperature superconducting (Hg-HIS, mainly Trig-1212 of  $T_c \sim 125$  K and Hg-1223 of  $T_c \sim 135$  K) thin films on metal substrates. Four students were partially supported during the reporting period. Research on the two research objectives has been carried out in parallel and exciting progress has been made, as summarized in the following. Details of our results may be found in 1 book chapter, 28 papers (24 published/accepted, and 4 submitted), 1 US patent awarded and 2 patent disclosures submitted. In addition, two Ph.D. students graduated during the project period. Dr. Yiyuan Xie graduated in March of 2002 and joined IGC immediately afterwards and Dr. Roberto Aga, in December 2002 with honor, and now stay as a postdoctoral research at the University of Kansas.

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# ***Coating Conductors with the Highest- $T_c$ Hg-based Superconductors***

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Final Report

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## **Summary**

This project started on April 1, 2000 and was completed on April 30, 2003. The overall goal of the project is to develop novel processes, such as the cation exchange process, for epitaxy of Hg-based high temperature superconducting (Hg-HTS, mainly Hg-1212 of  $T_c \sim 125$  K and Hg-1223 of  $T_c \sim 135$  K) thin films on metal substrates. Four students were partially supported during the reporting period. Research on the two research objectives has been carried out in parallel and exciting progress has been made, as summarized in the following. Details of our results may be found in 1 book chapter, 28 papers (24 published/accepted, and 4 submitted), 1 US patent awarded and 2 patent disclosures submitted. In addition, two Ph.D. students graduated during the project period. Dr. Yiyuan Xie graduated in March of 2002 and joined IGC immediately afterwards and Dr. Roberto Aga, in December 2002 with honor, and now stay as a postdoctoral research at the University of Kansas.

## **Major experimental results**

Under the AFOSR supports, the KU group has actively worked on coated conductors during the past few years. One major progress is in development of novel processes for epitaxy of the Hg-HTS films and coated conductors (three US patents awarded and one pending). Hg-HTSs have  $T_c$ s up to 135 K, the highest among HTSs, and intrinsically high irreversible fields with  $H_{irr}(77K) \sim 4-5$  T and  $H_{irr}(100K) \sim 1$  T. The higher  $T_c$ s allow higher operation temperatures, which implies significantly reduced cryogenic requirements and operation costs. In fact, the cooling requirement may be eliminated for many space-related applications as the background temperature in deep space is  $\sim 80$  K. On Hg-HTS thin films grown on single-crystal  $\text{LaAlO}_3$  substrates, we have achieved  $J_c$  values near  $2 \text{ MA/cm}^2$  at 100K and self field, which are about two orders of magnitude higher than those of any existing superconductors. These films have superior microwave properties and prototype microwave devices are under development in collaboration with the Superconductor Technology, Inc. Through collaboration with ORNL, a  $J_c(100K) \sim 1 \text{ MA/cm}^2$  has also been demonstrated on Hg-1212 coated  $\text{CeO}_2/\text{YSZ}/\text{CeO}_2/\text{RABiTs}$  tapes. This makes Hg-HTS coated conductors very appealing for power-related applications at 100 K or higher.

Our AFOSR project had two major focuses: developing processes to scale up Hg-1212 coated conductors, and exploring ways to improving the magnetic pinning strength of them. For the former, we have developed a batch processing technique and demonstrated that uniform Hg-1212 coated conductors over 10 cm Buffered Ni substrates. This technique can be applied for longer tapes when substrates are available. In collaboration with ORNL and LANL, we have shown that the magnetic flux pinning properties can be significantly improved on Hg-HTS films when linear defects are introduced either using GeV ion beam irradiation or substrate engineering. The latter is extremely appealing since it can be scaled up for long length processing. The  $H_{irr}$  of Hg-1212 and Hg-1223 has been increased to about 6T at 77 K and ~3 T at 100 K, adequate for many power-related applications at temperatures near and above 77 K.

**Objective 1: Research on fundamental issues related to and development of schemes for scale-up of Hg-HTS coated conductors**

- **Large area**—Many applications require large-area thin films. Synthesis of Hg-HTS materials requires a high vapor pressure of Hg in the range of 5-10 atm. To generate and maintain such pressures, the precursor samples of desired nominal composition have to be torch sealed with the Hg-vapor source (e.g. HgO, HgF<sub>2</sub>, HgCl<sub>2</sub>, HgCaO<sub>2</sub>, etc) in evacuated quartz ampoules for heat treatments to form Hg-HTS materials. We recently developed a reusable crucible technique that allows epitaxy of Hg-HTS films in a dynamic equilibrium of Hg and oxygen vapors, instead of a static equilibrium as in the sealed quartz ampoule. In addition, the crucible can be reused repeatedly and has no size restriction. In a preliminary test, we have obtained samples of quality comparable to that from in the sealed quartz ampoule technique. This technique is a very important step in scale up the Hg-HTS films to large area for various applications in both electronics and electrics. Our future effort will be in optimizing each step and finalizing the design of the crucible.
- **Long length**—HTScc must have long length for applications. We recently developed a batch process for scaling up Hg-HTS coated conductors (Hg-HTScc). The precursor tapes are wound around a source cylinder that is to provide either Tl or Hg vapor when heated in a commercial tube furnace. We have demonstrated that uniform physical properties (XRD,  $T_c$  and  $J_c$ ) can be obtained on Tl-HTS and Hg-HTC coated conductors of several cm long. For example, the end-to-end  $J_c$  (77 K) in exceeding 1 MA/cm<sup>2</sup> has been obtained on 2-cm long Hg-1212/Ni tapes. Experiments on longer tapes are underway in collaboration with doe labs and our focus will be to optimize the process for long tape fabrication.

**Objective 2: Study of vortex dynamics and optimization of pinning strength in Hg-HTS coated conductors**

One of the Air Force specifications for HTS coated conductor applications is the high irreversibility field  $H_{irr}$  in the range of 2-5 T at 77K. Hg-HTS has its original  $H_{irr}$  ~ 2-3 T at 77 K so improvement of  $H_{irr}$  is necessary. Last year, we demonstrated that the  $H_{irr}$  of Hg-1212 film is significantly improved with introduction of linear defects along the c-axis using GeV ion beam irradiation. For example, the  $H_{irr}$  was increased to ~ 6T at 77K and ~ 3T at 100K. This year's focus

has been on exploring practical ways to improve the  $H_{irr}$  and two schemes turned out to be successful: oxygen overdoping and substrate engineering. Details will be given in the following.

- **Oxygen Overdoping**—HTS's properties are sensitively determined by the oxygen content and the best  $J_c$  and  $H_{irr}$  are obtained when samples are moderately overdoped. For Hg-HTS, we developed a simple Fluorine-assisted process that allows oxygen overdoping in Hg-HTS at atmospheric pressures. The  $J_c$ s of overdoped Hg-1212 films were increased fairly uniformly over a wide temperature range by a factor of 2 and the  $H_{irr}$  significantly. For example, at 77 K,  $H_{irr}$  is increased by more than 50%, and this improvement is more significant at lower temperatures. The future research will be on understanding the mechanism of this Fluorine-assisted process. In addition, overdoping grain-boundaries will also be investigated using this technique.
- **Substrate Engineering**—HTS films show different microstructures when grown on different substrates. Linear growth defects could be formed when there is additional strain generated in film when substrate has a small vicinal angle with respect to the normal of the substrate. These linear defects may mimic the role of those generated by GeV ion beam irradiation, resulting in improved  $H_{irr}$ . On 4° STO vicinal substrates, we demonstrated recently that both in-field  $J_c$  and  $H_{irr}$  are improved. For example, at 77 K and  $H = 2T$ ,  $J_c$  is one order of magnitude higher in Hg-1212 film on 4° STO vicinal substrate than the same film on flat STO substrates. The  $H_{irr}$  at 77 K is enhanced by more than 30% on the former. The ongoing research is study the effect of vicinal angle on the  $J_c$ s and  $H_{irr}$  so as to optimize the improvement. In addition, we are exploring ways to generate vicinal buffers so that improved  $J_c$ s and  $H_{irr}$  can be obtained on coated conductors.

In addition to research on growth of coated conductors, we have also worked on development of appropriate tools for coated conductors characterization (partially from our AFOSR/MURI support). Scanning probe microscopy (SPM) has been a useful tool [20] for this purpose but most commercial SPM measures one physical property at a time. This is in sharp contrast to the need to correlate different physical property at a nanometer scale in most practical research and applications. Motivated by such needs to correlate different physical properties at a microscopic scale, we have recently developed a dual-channel scanning probe microscope (a US patent pending). The novel feature of this invention is that it uses a coaxial multi-layered probe design, with each layer (made using high-vacuum thin film coating process) serves as a channel for different input/output signal. Since the layer thickness is in the nanometer regime, this multi-layered probe allows imaging/mapping of two or more different properties at the same pixel simultaneously. For example, an optical fiber tapered to few tens of nanometers in diameter can be used for transferring optical signals. Coating this fiber with a thin layer of metals (Al, Au, etc) will make the fiber electrically conducting. If this fiber is used as the center conductor of a  $\frac{1}{4}$  wavelength coaxial resonator, both optical and microwave signals can be transferred through the probe. This near-field scanning optical/microwave dual-probe microscope (NSOM/NSMM) has been demonstrated experimentally in PI's lab recently [21] and will provide a unique tool for mapping physical properties of the coated conductors

## **Publications**

### **Patents:**

1. (Patent Disclosed) R. Aga and J.Z. Wu, "Development of Near-field Scanning Optical/microwave Dual Probes", submitted Nov. 15, 2001.
2. (Patent Disclosed) Y.Y. Xie and J.Z. Wu, "Re-usable crucible technique for large area epitaxy of Hg-based high-temperature superconducting wafers and long-length tapes", submitted Oct. 18, 2001.
3. U.S. patent 6395685 (2002), J.Z. Wu, S.L. Yan, and Y.Y. Xie, "Thin Film Hg-Based Superconductors and Method of Fabrication Thereof".

#### **Ph.D. Thesis:**

21. R. Aga, Jr., honorable thesis, "Development of Dual-channel Scanning Optical/microwave microprobe microscopy", Department of Physics and Astronomy, University of Kansas, December 2002.
22. Y.Y. Xie, "Epitaxy of Hg-based high temperature superconducting films on oxide and metal substrates", Department of Physics and Astronomy, University of Kansas, May 2002.

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2. Y.Y. Xie, J.Z. Wu, S.H. Yun, R. Emergo, and R. Aga, "Magnetic flux pinning enhancement in vicinal  $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$  films", submitted to Appl. Phys. Lett.
3. R. Aga and J.Z. Wu, "Near-field Scanning Optical/microwave Dual Probe", submitted to Appl. Phys. Lett.
4. Z.W. Xing and J.Z. Wu, "Reversible cation exchange in "1212" superconductors", submitted to Appl. Phys. Lett.
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